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## Magnetic phase transitions in $R_2Ni_3Si_5$

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### Abstract

Neutron diffraction has been used to investigate the magnetic and crystal structures in  $R_2Ni_3Si_5$  ( $R = Pr, Nd, Tb, Dy, Ho$ ). Our high-resolution data indicate that this system crystallizes in the *Ibam* space group. *Pr* orders antiferromagnetically at  $T_N = 8.8$  K, with a magnetic unit cell that is double the chemical unit cell along the *b* direction. The magnetic and chemical unit cells are the same for the *Nd* spins, which order at 9.6 K. *Tb* first orders with an incommensurate modulated structure at 19 K, and then changes to a commensurate structure at 12.2 K. An incommensurate structure is also observed for *Dy*, which orders at 9 K, with a lock-in transition at  $\sim 4$  K. The *Ho* system has an incommensurate structure with an ordering temperature of 6.9 K. © 1998 Published by Elsevier Science B.V. All rights reserved.

**Keywords:** Magnetic order; Neutron diffraction; Magnetic transitions

The  $R_2Ni_3Si_5$  ( $R$  – rare earth) system exhibits, very interesting magnetic and transport properties [1–5]. For example, magnetic susceptibility and specific heat measurements in some of these compounds indicate double magnetic transitions. Recently, giant magnetoresistance (MR) and anomalous MR behavior have also been observed in these materials, and have been interpreted in terms of rare-earth magnetic interactions and magnetic

structures (as *Ni* carries no significant moment). In order to understand the nature of the magnetic transitions in this class of compounds, we have carried out neutron diffraction experiments on  $R_2Ni_3Si_5$  for  $R = Pr, Nd, Tb, Dy, Ho$ . The rare earth spins in all these materials order antiferromagnetically with a wide variety of both commensurate and incommensurate magnetic structures, while additional transitions associated with a change in magnetic structure are observed in *Tb, Dy* and *Ho* compounds.

Neutron diffraction experiments were conducted on polycrystalline samples on a triple-axis spectrometer in two-axis mode at the NIST Center for Neutron Research. A pyrolytic graphite

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monochromator and filter were employed at a wavelength of 2.351 Å. High-resolution profile refinements were carried out with data taken on the BT-1 spectrometer to characterize the sample at room temperature. The details of the sample preparation technique can be found elsewhere [1–5]. For the low-temperature measurements (down to 1.4 K), a pumped  $^4\text{He}$  cryostat (ILL) was used.

Room-temperature refinements showed that all these samples were of single phase. The nuclear peaks can be indexed as  $(h, k, l)$  with  $h + k + l = \text{even}$ , and our analysis indicates that  $\text{R}_2\text{Ni}_3\text{Si}_5$  crystallizes in the orthorhombic *Ibam* space group in agreement with the previous X-ray analysis [1–6]. The lattice parameters and the unit cell volume were found to increase with increasing lanthanide radius as expected. There is no indication of any structural transition in any of these samples down to 1.8 K. Details of the crystal and magnetic structures will be presented elsewhere [7].

For the  $\text{Pr}_2\text{Ni}_3\text{Si}_5$  and  $\text{Nd}_2\text{Ni}_3\text{Si}_5$  materials commensurate antiferromagnetic magnetic structures are observed. The Pr spins order at 8.8 K, and all the observed magnetic peaks can be indexed as  $(h, k + \frac{1}{2}, l)$  based on the chemical unit cell with  $h + k + l = \text{even}$ . This indicates that the magnetic unit cell is double the chemical unit cell along the *b*-direction, while it is the same in the other two directions. The low-*T* ordered magnetic moment of  $2.1(1)\mu_{\text{B}}$  is directed along the *b*-axis. In the Nd compound the magnetic peaks can be indexed as  $(h, k, l)$  with  $h + k + l = \text{odd}$ , and therefore, the magnetic and chemical unit cells are the same for  $\text{Nd}_2\text{Ni}_3\text{Si}_5$ . The Néel temperature is 9.6 K.

Two magnetic transitions are observed in  $\text{Tb}_2\text{Ni}_3\text{Si}_5$ , and a portion of the magnetic diffraction pattern is shown in Fig. 1. The Tb moments order at  $T_{\text{N}1} = 19\text{ K}$  in an incommensurate structure with a wave vector  $\delta = (0, 1.0, 0.2055)$ . This structure abruptly locks-in to a commensurate structure [ $\delta = (0, 1.0, 0.0)$ ] at  $T_{\text{N}2} = 12.2\text{ K}$ . We observed  $\sim 0.25\text{ K}$  temperature hysteresis near  $T_{\text{N}2}$  while near  $T_{\text{N}1}$  the data are the same on cooling and warming. Our results indicate the Tb magnetic moment is along the *c*-direction, in agreement with a recent report [8].

The Dy material orders at  $T_{\text{N}1} = 9\text{ K}$  with a structure that is also incommensurate, and the

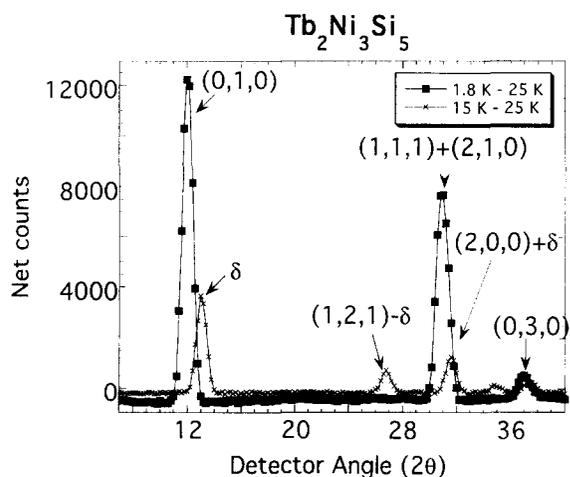


Fig. 1. Magnetic diffraction pattern for  $\text{Tb}_2\text{Ni}_3\text{Si}_5$  obtained by subtracting the high-temperature data (25 K) from the low-temperature data (1.8 and 15 K).

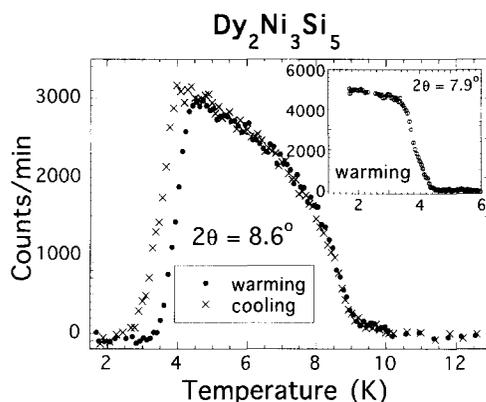


Fig. 2. Temperature dependence of the magnetic Bragg peak at  $2\theta = 8.6^\circ$  for  $\text{Dy}_2\text{Ni}_3\text{Si}_5$  on warming and cooling. The inset shows the temperature dependence of the magnetic Bragg peak at  $2\theta = 7.9^\circ$ .

temperature dependence of one of the magnetic peaks is shown in Fig. 2. The intensity increases with decreasing temperature below  $T_{\text{N}1}$ , and then drops to zero at  $T_{\text{N}2} = 3.5\text{ K}$  due to a lock-in transition to a commensurate structure with  $\delta = (0, 1, \frac{1}{3})$ . Therefore, the magnetic unit cell is triple the chemical unit cell along the *b*-direction, while it is the same in the other two directions. The temperature dependence of the  $(0, 1, \frac{1}{3})$  magnetic

intensity is shown in the inset of Fig. 2, and clearly indicates the transition at  $T_{N2}$ . Fig. 2 also shows  $\sim 0.5$  K hysteresis at  $T_{N2}$ , while no hysteresis is observed at  $T_{N1}$ . In addition, there is an indication of another magnetic phase in a narrow temperature region ( $\sim 1$  K) near  $T_{N2}$ .

Like Tb and Dy, the Ho system shows a double transition, but in this case both magnetic structures are incommensurate. The Ho moments order antiferromagnetically at 6.8 K, and the magnetic structure starts to change below 2.2 K.

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